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MONITORING OF COW'S MILK FOR FRESH FISSION PRODUCTS
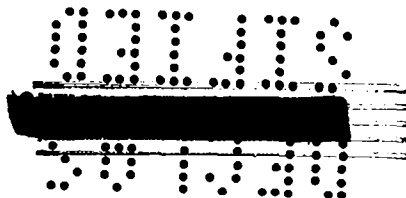## FOLLOWING AN ATOMIC DETONATION

by

William S. Johnson
Jean McClelland
Clarence P. Skillern

HEALTH AND SAFETY

ABSTRACT

Milk samples were collected on an approximately daily basis from various points both in and out of the fall-out pattern resulting from Shot Harry, Upshot-Knothole Operation, detonated on 19 May 1953. Sample collection was limited generally to the purchase of bottled or packaged milk from the retail market. An analytical procedure with good recoveries was developed to determine the activity in the milk. The concentrations found, when compared to an emergency tolerance value for water, do not represent in themselves a source of internal hazard. The data presented tend to indicate that the principal source of contamination was from material ingested by the animals.

ACKNOWLEDGMENT

The writers wish to express their appreciation to the members of the Off-Site Radiological Safety Group who participated in the operational phase of this investigation at the Nevada Proving Grounds.

Following the delineation of the off-site fallout pattern associated with Shot Harry of the Upshot-Knothole Series at the Nevada Proving Grounds, interest was expressed in the possibility of fission-product contamination of milk originating in the St. George, Utah, milkshed. Figure 1 is a map of the fallout pattern from Shot Harry and the area involved. The Off-Site Group of the Radiological Safety Organization, NPG, was assigned to the field investigation. This report sets forth the information obtained from this investigation with those conclusions which are apparent to the authors.

On confronting this problem it was found that no standard procedure existed for preparing milk samples for counting and that there were no tolerance levels for fission-product contamination of milk. Since the collection of samples could not be delayed until these problems were solved, the investigation was initiated on D+1 day following Shot Harry. This phase of the program is incomplete since no details were obtained on time of milking, milking technics, individual producers, etc. Off-Site personnel in the St. George area decided that extensive inquiry into such details would indicate the concern of the test program's rad-safe group with the possibility of milk contamination and alarm an already-worried community. Subsequent events have proved the wisdom of this decision. Therefore the milk collection was limited primarily to purchases of bottled or packaged milk from local retail outlets on approximately a daily basis. The Off-Site monitor at St. George was able to determine that the first sample purchased (5/21/53 AM) very probably came from a milking on the evening of D-day. Thus a lag of 24 to 36 hours between time of milking and appearance on the retail market may be assumed. At the same time the Los Alamos Scientific Laboratory was developing a method for preparing milk samples for counting. Further work done at NPG resulted in the procedure outlined in Appendix A.

To check the loss of activity in ashing, transferring, and plating, a solution of fission products was prepared by ashing and extracting a filter paper sample of known activity. Two 100 ml aliquots of milk, known to come from an area other than St. George, were spiked with 5 ml of the fission-product solution. These aliquots were treated in the same manner as the field samples. Another 5 ml aliquot was plated directly and counted at the same time as the spiked milk in order to eliminate error due to decay. Table 1 shows recoveries of 96 to 98%. The small correction factor is not included in the reported results.

The counting planchet used for all milk samples was a rectangular piece of aluminum foil 1.5 mil thick with a 4" x 9" counting surface and 1/8" to 1/4" sides to contain the solution (Figs. 2 and 3). The size of the planchet was determined by the maximum dimensions of the window of the counting probe which was available. A planchet of this size reduced counting loss due to self-absorption by the residue of inorganic salts. Counts were made for 2 minutes or longer with a methane-flow proportional counter probe having a window thickness

of approximately 0.75 mg/cm^2 and the counts were then compared to a standard Sr^{90}-Y^{90} beta solution plated on a clean planchet. The counter efficiency was found to be approximately 40%. Normal background with a blank planchet in place was generally from 1000 to 1500 c/m depending on the amount of activity being counted on adjacent instruments. The background count was determined at the beginning and end of each series of counts and often between samples.

The concentration of beta activity in each sample at the time of its first count is given in Table 2. Due to the lack of an analytical procedure, the first count was not obtained until D+6 days. It is significant to note that five samples contained no detectable activity at the time of the first count, hence the activity found was not naturally occurring. All samples showing activity were followed for decay characteristics. With the exception of Sample #1 the activity soon dropped to a low level which gave poor counting statistics and erratic results. The decay of Sample #1 is plotted in Fig. 4 with that of an air sample collected in St. George during the period of maximum fallout. A line of -1.2 slope represents the theoretical decay.[1] The deviation of the air sample decay from a straight line in the vicinity of 4 to 5 days has been reported.[2] Since the early portion (6 to 8 days) of the milk sample decay approximates the decay of the air sample, the assumption is made, for the purposes of extrapolation, that the same decay occurred in the milk. The fission-product concentrations have been extrapolated to 0800 on the date of purchase, which is assumed to be the time of maximum concentration available to the consumer. Table 3 was prepared on the basis of these arbitrary assumptions. In the absence of a specific tolerance level for milk contaminated with fission products, the proposed emergency value for water is used for comparison. This is shown in the last column of the table. The maximum permissible concentrations given are those of Morgan and Straub[3] where $MPC = Kt^{-1.2}$ In this equation K = 1 and t is days after detonation. This gives concentration in microcuries per liter. The maximum permissible concentration for continuous consumption is obtained by the use of this equation at its lower limit, i.e., $t \approx 10^4$ days. The accepted value for this is given as 1×10^{-4} μc/liter in Handbook 52.[4]

1. Los Alamos Scientific Laboratory, The Effect of Atomic Weapons, U.S. Government Printing Office (1950).

2. W. S. Johnson, H. F. Schulte, and E. C. Hyatt, "Report of the Advisory Personnel to the Air Sampling Program," WT-566 (in process 1953). Confidential.

3. K. Z. Morgan and C. P. Straub, "External and Internal Exposure to Ionizing Radiation and Maximum Permissible Concentration (MPC) of Radioactive Contamination in Air and Water Following an Atomic Explosion," AECU-2332 (April 1952).

4. National Bureau of Standards, Handbook 52, Maximum Permissible Amounts of Radioisotopes in the Human Body and Maximum Permissible Concentrations in Air and Water, U. S. Government Printing Office (1953).
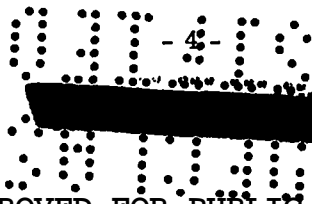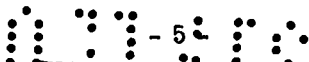
Obviously any hazard associated with fission-product contaminated milk will be limited in time to the early region of the decay curve governed by the equation MPC $= Kt^{-1.2}$ (i.e., emergency values). Comparisons to emergency values would be more realistic than comparisons to the level for continuous exposure. On this basis it is apparent that, although significant quantities of activity were found in the milk samples collected, they did not represent a source in themselves of an internal hazard.

Because of the lack of knowledge of the history of the milk samples prior to purchase it is relevant to consider the mode of entry of the contamination found in the milk. The possibility of contamination occurring at any or all points in the milk production should be considered. Conclusive evidence would be obtained only from a controlled investigation of these particular facets of the problem. However, there are pertinent points which may be derived from the data obtained to support a contention that the reported activities could be present in the milk as it is drawn from the cow.

The rapid change in slope of the milk sample decay curve in Fig. 4 at about D+10 to 12 days would indicate that the activity is from something other than gross fission products. Two other samples (#2 and #12), which are not presented graphically, indicate a similar sharp deviation at this time. On the assumption that a selective absorption of certain fission isotopes has occurred within the animal, this curve would then represent the resultant decay scheme of a gross fission sample less the contributions of the absorbed isotopes. If the contamination has entered the milk during or after the milking procedure its decay characteristic should be that of a gross fission-product sample. In addition a goat milk sample was collected under the observation of a monitor. A one-half pint jar of unknown history was filled with milk directly as it came from the animal in an open yard. No attempt was made during or following the operation to prevent contamination as would have been the case with technics used by a Grade A milk producer. Thus it might be expected that the contamination found could arise from several sources. Although the low activity of this sample increases the possibility of counting error, at the time of criticality of Sample #1 the decay curve of #7 is dissimilar (Fig. 4). This curve follows the $t^{-1.2}$ law indicating the probability of external rather than internal contamination in this case.

TABLE 1

RECOVERIES OF ANALYTICAL PROCEDURE FOR MILK

Sample	Net D/M	Recovery (%)
5 ml of fission-product solution	34,200	
5 ml of fission-product solution in 100 ml of milk	33,800	98.8
5 ml of fission-product solution in 100 ml of milk	33,080	96.7

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TABLE 2

CONCENTRATION OF FISSION PRODUCTS IN COLLECTED MILK SAMPLES

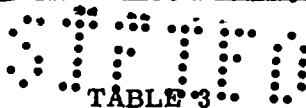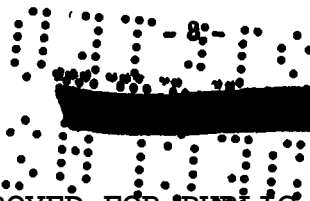| Sample No.	Sample Data	Time of Count	Net D/M/100 ml	μc/liter at Time of Count
1	St. George Ice Co. Grade A, pasteurized, purchased 5/21/53 AM, probably 5/19/53 PM milking.	1000 5/25/53	18,892	85.9×10^{-3}
2	St. George Ice Co. Grade A, pasteurized, purchased 5/22/53.	1010 5/25/53	3,460	15.7×10^{-3}
3	Anderson Dairy, Las Vegas Nev., Grade A, vitamin D, purchased in Las Vegas 5/22/53. St. George producers supply Anderson with some of its milk.	1015 5/25/53	4,620	21.0×10^{-3}
4	Clark Dairy, Las Vegas, Nev., homogenized, vitamin D, purchased in Mesquite, Nev., 5/22/53. Clark Dairy supplied by producers in Mesquite and Bunkerville.	1025 5/25/53	Background	
5	Farm of Max Hafen, Mesquite, Nev., from milking 5/19/53 PM.	1030 5/25/53	Background	
6	Farm of Max Hafen, Mesquite, Nev., from milking 5/20/53 PM.	1035 5/25/53	Background	
7	Goat milk from Mill's Farm, LaVerkin, Utah (3 miles north of Hurricane, Utah), from milking 5/21/53 AM.	1600 5/25/53	1218	5.5×10^{-3}
8	Clark Dairy, homogenized, vitamin D, purchased in Las Vegas 5/24/53.	1900 5/25/53	Background	
9	St. George Ice Co. Grade A, pasteurized, purchased 5/23/53.	1910 5/25/53	1922	8.7×10^{-3}
10	Clark Dairy, homogenized, vitamin D, purchased in Las Vegas 5/26/53.	2100 5/27/53	Background	
11	St. George Ice Co. Grade A, pasteurized, purchased 5/25/53.	2105 5/27/53	2282	10.4×10^{-4}
12	St. George Ice Co. Grade A, pasteurized, purchased 5/26/53.	2115 5/27/53	2995	13.6×10^{-4}
13	St. George Ice Co. Grade A, pasteurized, purchased 6/1/53.	2020 6/ 3/53	1455	6.6×10^{-4}
14	St. George Ice Co. Grade A, pasteurized, purchased 6/2/53.	2025 6/ 3/53	1927	8.7×10^{-4}
15	St. George Ice Co. Grade A, pasteurized, purchased 6/3/53.	2035 6/ 3/53	2950	13.4×10^{-4}

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TABLE 3

CONCENTRATION OF FISSION PRODUCTS IN COLLECTED MILK SAMPLES

AT ASSUMED TIME OF MAXIMUM ACTIVITY

Sample No.	Date of Purchase	μc/liter at 0800 on Day of Purchase	Maximum Permissible Concentration (μc/liter)*
1	5/21/53	459×10^{-3}	405×10^{-3}
2	5/22/53	60×10^{-3}	255×10^{-3}
3	5/22/53	75×10^{-3}	255×10^{-3}
7	5/21/53	30×10^{-3}	405×10^{-3}
9	5/23/53	23×10^{-3}	182×10^{-3}
11	5/25/53	16×10^{-3}	114×10^{-3}
12	5/26/53	17×10^{-3}	95×10^{-3}
13	6/ 1/53	8×10^{-3}	46×10^{-3}
14	6/ 2/53	10×10^{-3}	42×10^{-3}
15	6/ 3/53	14×10^{-3}	38×10^{-3}

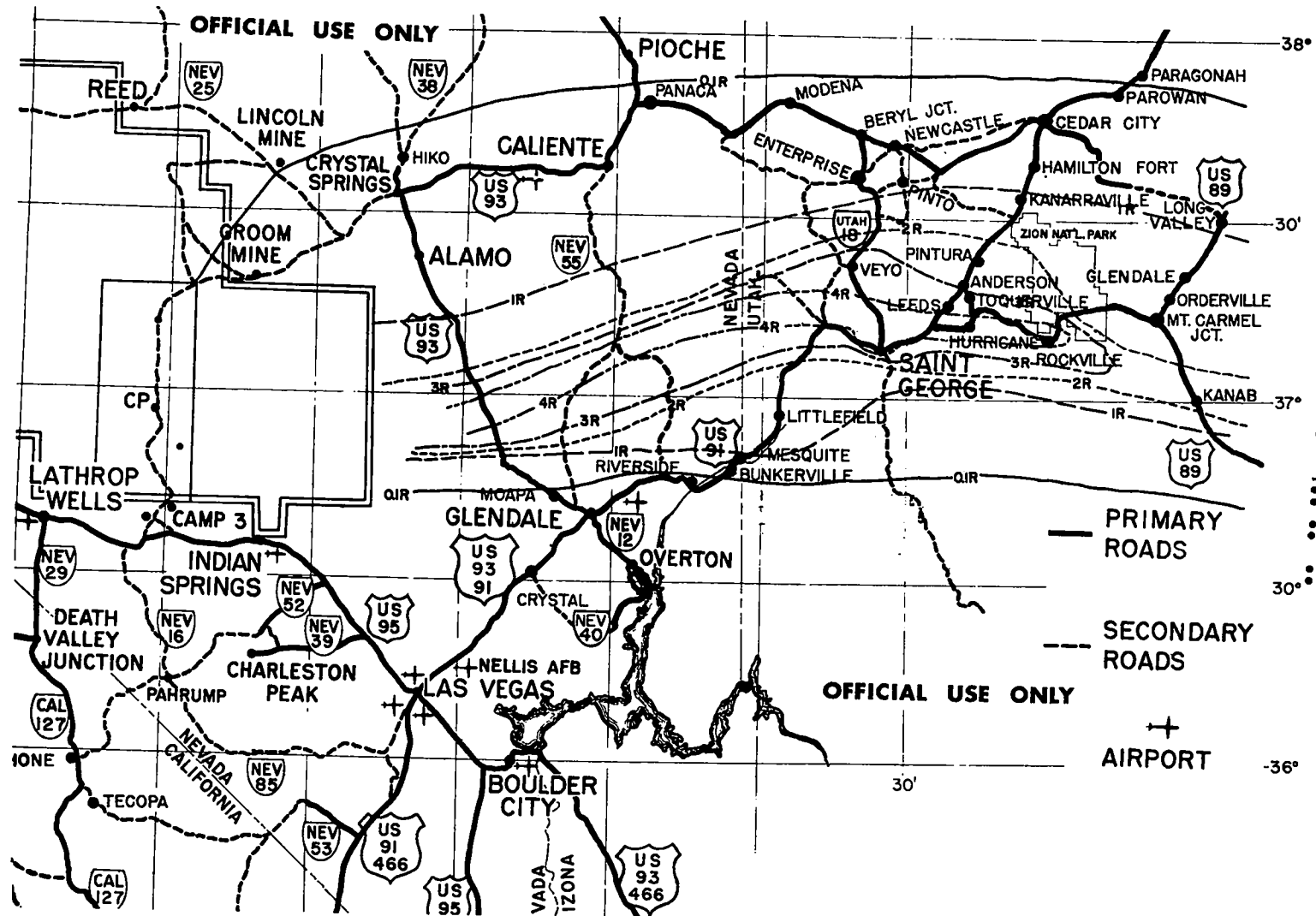* The MPC is calculated for the same time as the preceding column, or 0800 on the day of purchase, using MPC $= Kt^{-1.2}$.

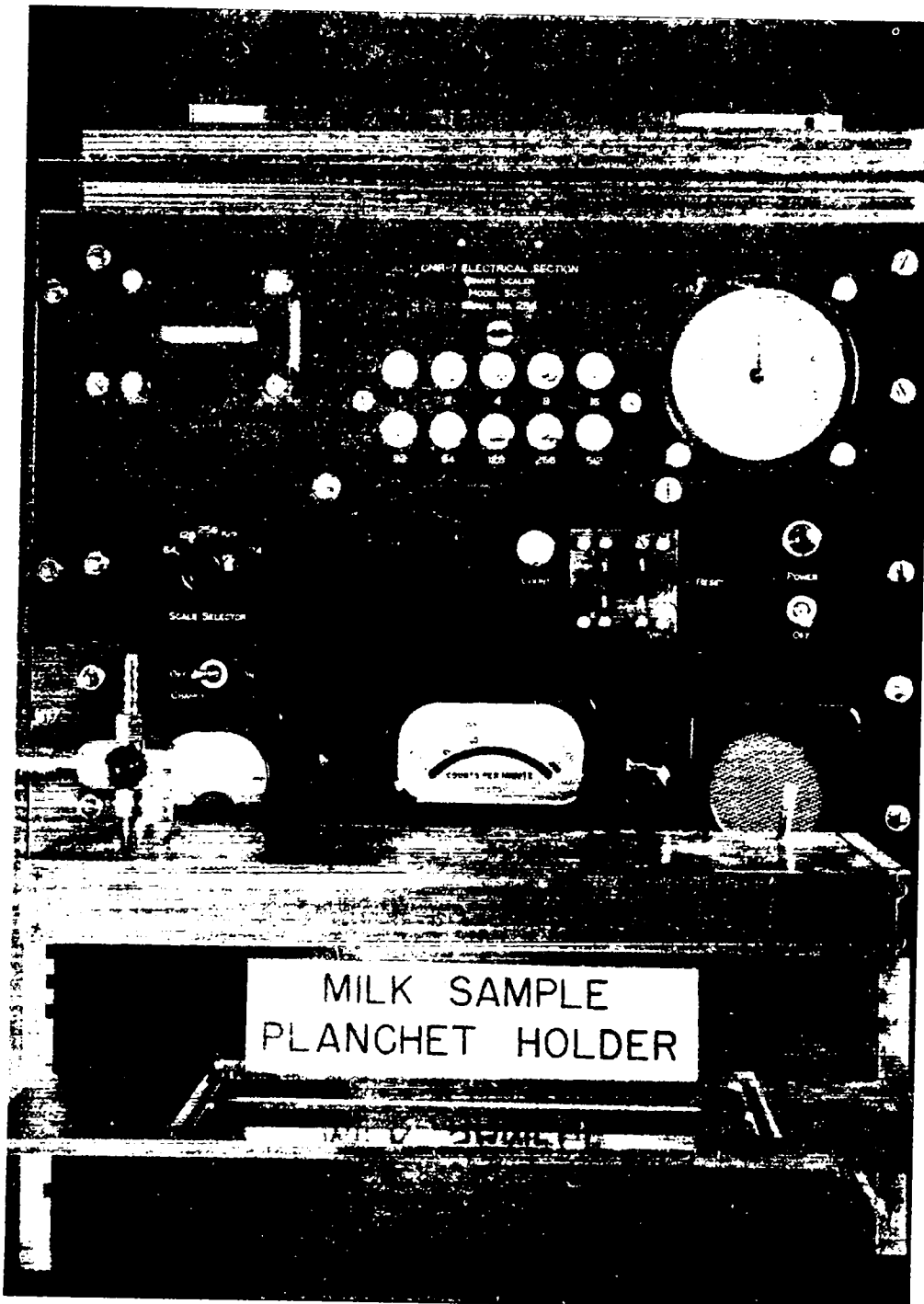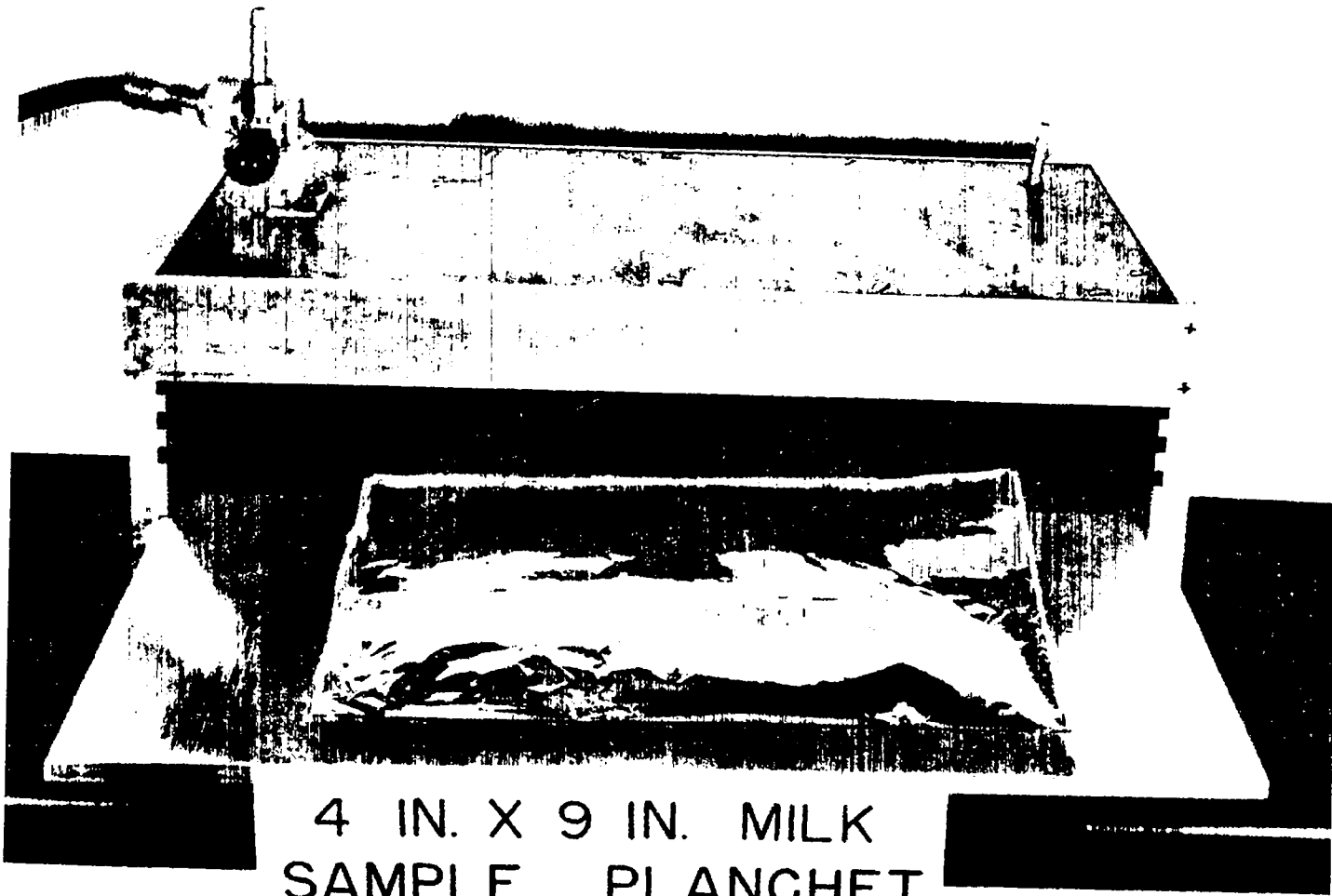Fig. 1. Seventy year isodose plot of fallout from Shot Harry. Time of
Shot—0505 PDT, 19 May 1953.

Fig. 2. Photograph of counter, probe, and planchet holder.

4 IN. X 9 IN. MILK
SAMPLE PLANCHET

Fig. 3. Photograph of planchet and probe.

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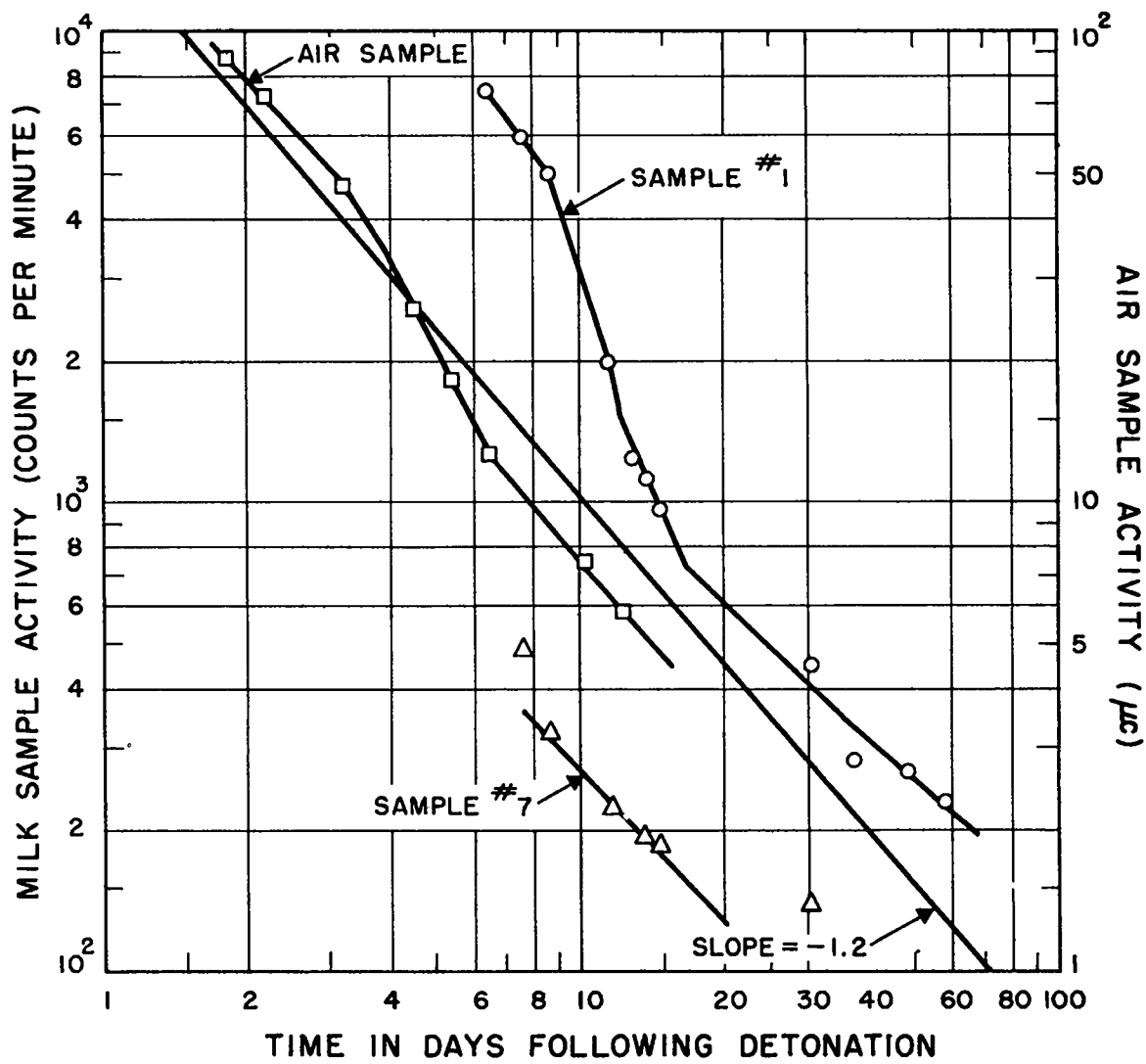Fig. 4. Decay curves of various samples from Shot Harry.

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APPENDIX A

LABORATORY PROCEDURES FOR MILK SAMPLES

1. Shake or stir sample thoroughly and transfer 100 ml to a 600 ml beaker for slow evaporation. Mark the beaker with a code symbol for the sample with a wax glass-marking pencil.

2. Add 25 ml concentrated nitric acid and a pinch of carborundum boiling chips to each beaker. Cover with a speedy-vap.

3. Evaporate the sample to dryness on a hot plate at low heat.

4. To the residue, add 20 ml concentrated nitric acid and again take to dryness, using high heat. Repeat this until the ashed salts are white, taking care to rinse down the sides of the beaker with concentrated nitric acid. The speedy-vap may be dispensed with after the second or third time. Cool the beaker before each addition of acid. The sample will probably have to be taken to dryness 5 or 6 times before the salts are white and the organic matter completely destroyed.

5. To the white ashed salts, add 2 ml of concentrated nitric acid and 30 ml of water. To this solution add 5 ml of hydrogen peroxide and boil until all the hydrogen peroxide has boiled off and the volume is reduced to 5 to 10 ml.

6. Transfer the hot residue with a micro-pipette to a special planchet. Dissolve the residue on the sides of the beaker and that remaining in the bottom of the beaker with dilute (1:1) nitric acid. Transfer the washings to a planchet. Dry under an infrared lamp. If the volume is large, the planchet may be placed on a hot plate, turned on low, and covered with an asbestos pad, to hasten drying.

7. After the solution has been evaporated to dryness in the planchet, coat the planchet with Krylon (care must be taken to avoid blowing the salts off the planchet) and place in a properly labeled box to await determination of the activity of the residue.

8. Counting technics are the same as those employed for other samples including the use of a standard planchet for counter efficiency and a blank planchet for backgound determinations.

9. Report final result in μc/liter.